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**FINAL REPORT ON AFOSR-DARPA PROJECT**  
**Higher Performance (Nd,Pr)-(Fe,Co)-B Magnets**  
**(# AWD N0001H-00-1-0803)**

**PROJECT SUMMARY**

This project was focused on the advanced Nd(Pr)-Fe-B based magnets. The aim of this study was twofold: (1) to develop high performance nanocomposite  $R_2Fe_{14}B/\alpha$ -Fe magnets through adjustments in chemical compositions and processing conditions; and (2) to investigate the effect of small substitutions on the coercivity of the higher energy product  $(BH)_m$  Nd-Fe-B sintered magnets in an attempt to increase their coercivity without decreasing much their remanent magnetization. The studies on nanocomposite magnets have been very successful and led to the development of Pr-Tb-Fe-Nb-B magnets with  $(BH)_m=17.3$  MGOe. In the other project, Ga substitution was found to alter the intergranular morphology and chemical composition, and thus significantly increase the coercivity through domain wall pinning/nucleation.

**INTRODUCTION**

Many applications in automotive, information, and communication industries call for less-expensive permanent magnets with superior magnetic properties. One way to achieve this is to fabricate nanocomposite magnets consisting of a fine mixture of magnetically soft and hard phases (called "exchange-coupled magnets"). Other applications call for advanced magnets with higher operating temperature. The latter behavior can be obtained in sintered Nd-Fe-B magnets with minor element additions that lead to the formation of a special microstructure consisting of 2:14:1 grains, isolated by a very thin layer of a nonmagnetic phase. Our DARPA Project Contract (AWD N0001H-00-1-0803) was focused on both of these research areas. This report summarizes our major findings.

**(1) Nanocomposite magnets**

Melt-spun samples with composition based on  $(Pr,Tb,Dy)_8(Fe,Nb)_{86}B_6$  were studied systematically. Alloys were first prepared by arc melting under Ar atmosphere, and then melt-spun at different wheel speeds that were optimized for each composition. The obtained ribbons were crystallized with proper annealing to develop a fine nanoscale microstructure with optimum magnetic properties.

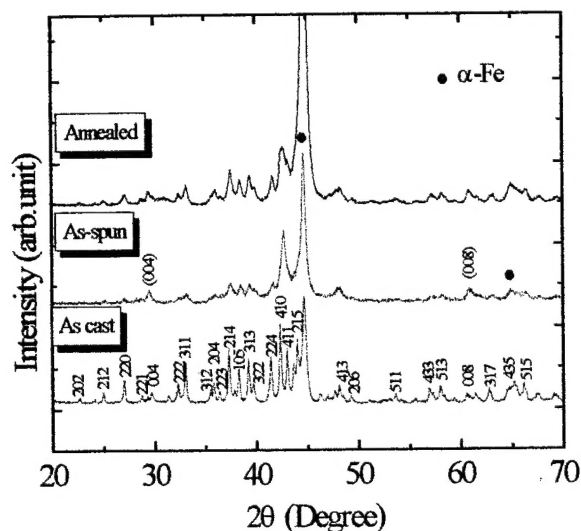


Fig. 1

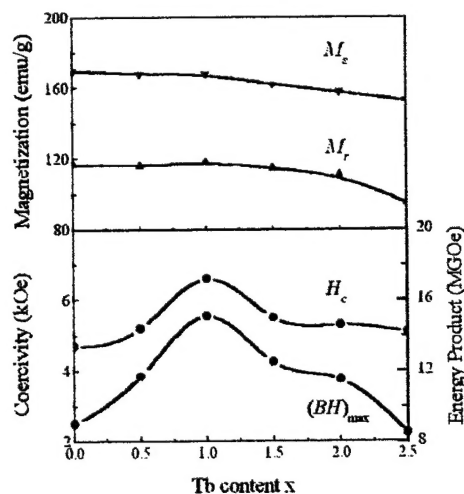


Fig. 2

The as-cast sample appears to have a mixed structure, consisting of the hard magnetic  $\text{Pr}_2\text{Fe}_{14}\text{B}$  and soft  $\alpha\text{-Fe}$  phases with large grain size as shown in Fig. 1. After melt spinning, the average grain size decreases. The optimally melt-spun ribbons show a fine nanocomposite structure, which results in an optimal coercivity. The studies also showed that the magnetic properties, including the coercivity, remanence and energy product, could be greatly improved with optimal heat treatment. Studies on the effects of small substitution showed that the coercivity first increases with the Tb and Dy content due to the enhancement of anisotropy, reaching a maximum of 6.5 kOe, and then decreases with further increase of the Tb and Dy content (Fig. 2). As a consequence, the maximum energy product goes through a peak of 15

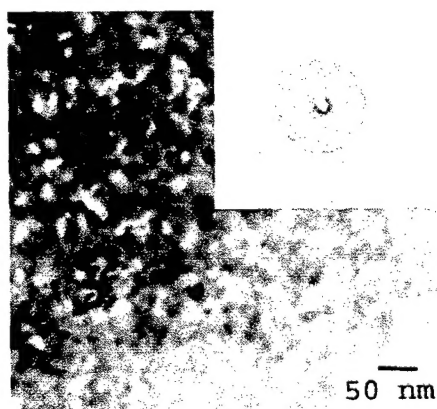


Fig. 3

MGOe with small additions as compared to 9.1 MGOe in (Tb, Dy)-free samples. Meanwhile, a microstructure with a much finer and uniform grain size was developed, leading to the increase of coercivity and energy product. Other substitutions of M ( $\text{M}=\text{Cr}$ , Nb, Ti and Co) for Fe have been investigated systematically. The results showed that Nb is the best element among all the substitutions investigated, that significantly reduces the grain size of  $\alpha\text{-Fe}$  and improves the

microstructure with a much finer and uniform grain size (Fig. 3). Moreover, Nb substitution leads to a lower wheel speed for the formation of 2:14:1 crystallites and a higher coercivity up to 9.8 kOe. The optimal overall properties obtained under optimum-quenching followed by a subsequent annealing are  $H_c = 8.6$  kOe and  $(BH)_m = 17.3$  MGOe in  $\text{Pr}_7\text{Tb}_1\text{Fe}_{87}\text{Nb}_{1.5}\text{B}_4$  as shown in Fig. 4, where the smooth demagnetization curve presents the typical characteristic of a single component system but with a high remanence value,  $M_r/M_s = 0.69$ . The coercivity and energy product are about twice higher than those in Nb-free samples.

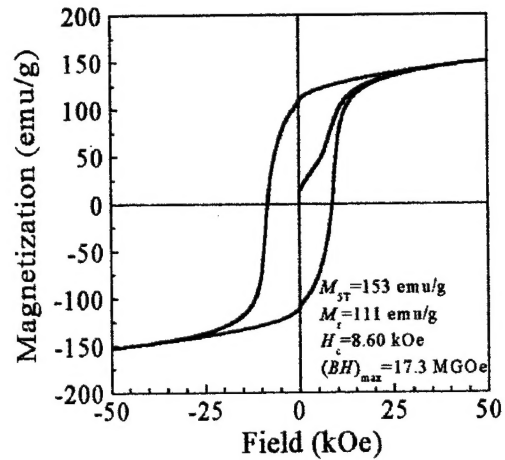


Fig. 4

## (2) Nd-Fe-B sintered magnets

To improve the temperature stability of Nd-Fe-B magnets, the effect of small substitutions on the coercivity of sintered magnets has been investigated. The tiny additions of Al, Cu, Mo, V and Sn to the Dy-free Nd-Fe-Co-B were found not to be effective for improving the coercivity. On the other hand, additions of Ga and Nb were found to be the most effective for drastically improving the room-temperature coercivity in both Nd-Fe-B and Nd-Fe-Co-B without decreasing much the remanent magnetization (Fig. 5). The effect of (Ga, Nb) additives on  $H_{ci}$  is

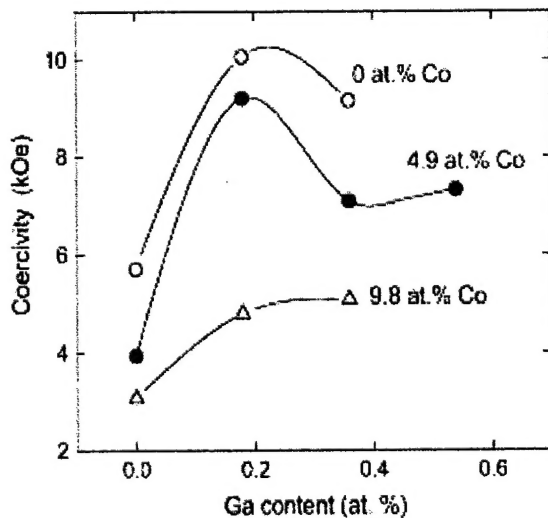


Fig. 5

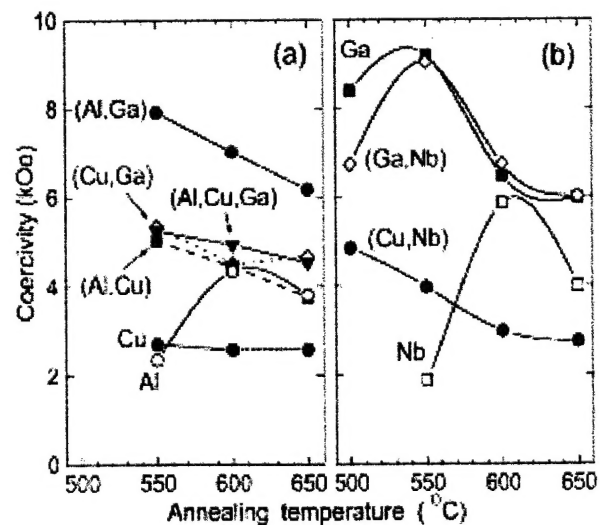


Fig. 6

equivalent to that of only Ga. It is more pronounced for magnets prepared by the powder blending technique. The Ga, (Al, Cu) and (Ga, Nb) additives decrease the optimum annealing temperature of the sintered magnets (Fig. 6). Scanning electron microscopy on Ga-free and Ga-added magnets did not reveal a noticeable difference in phase morphology (Fig. 7). In both magnets, the Nd-rich phase does not form continuous layers around the 2:14:1 grains and it appears to be mostly located in triple junctions. This morphology is typical for magnets with <15 at% Nd.

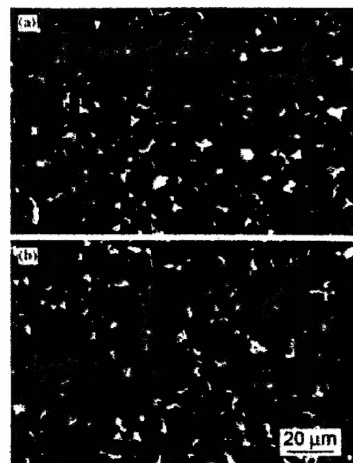


Fig. 7

However, Fe and Co concentrations in the inter-granular Nd-rich phase, as revealed using EDAX analysis (Table 1), were found to markedly increase with the small Ga addition. The combined addition of Co with a small amount of Cu did not increase the coercivity of the Dy-free magnets. However, none of small substitutions made can be considered as an alternative for Dy for extending the operating temperature range of the high-energy Nd-Fe-B magnets.

Table 1

|   | Treatment   | $H_c$ (kOe) | Nd rich phase |          |          |
|---|-------------|-------------|---------------|----------|----------|
|   |             |             | Nd (at%)      | Fe (at%) | Co (at%) |
| $\text{Nd}_{14.54}\text{Fe}_{74.2}\text{Co}_{4.9}\text{B}_{6.36}$                 | Annealed    | 4.0         | 85.5          | 12.6     | 1.9      |
| $\text{Nd}_{14.35}\text{Fe}_{74.2}\text{Co}_{4.9}\text{B}_{6.36}\text{Ga}_{0.18}$ | As-sintered | 5.4         | 88.8          | 9.6      | 1.6      |
| $\text{Nd}_{14.35}\text{Fe}_{74.2}\text{Co}_{4.9}\text{B}_{6.36}\text{Ga}_{0.18}$ | Annealed    | 9.2         | 69.8          | 19.3     | 10.9     |

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